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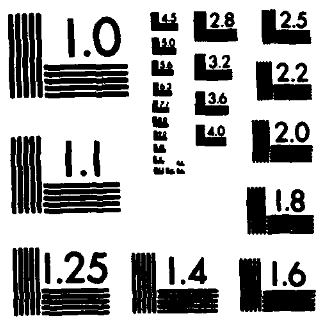

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DIFFRACTION FROM OVERLAYER ISLANDS WITH POSITIONAL CORRELATION

by

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Surface Science, in press

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Abstract

By representing the autocorrelation functions with a set of δ-functions in the method developed by Houston and Park, closed-form solutions are obtained for the angular distribution of diffracted intensity, in the kinematic approximation, from commensurately adsorbed overlayer islands with positional correlation. It is shown, for islands with unit mesh larger than that of the substrate, that the interference between the islands has only a minor influence on the angular distribution of the superlattice beam intensity if the islands have short-range correlations in their position. Although interference effects are present in the line shape for islands with long-range positional correlation, it is shown that these are not measurable unless a very narrow detector is used. The results are applicable to LEED, RHEED, and grazing-angle x-ray diffraction measurements.

I. Introduction

In adsorption on single-crystal surfaces, frequently two-dimensional ordered regions (islands) with a particular structure form at low coverages.[1] This is due to the existence of net attractive interactions between the adsorbate atoms.[2] For a perfect surface at equilibrium, the minimization of the free energy of the system requires that one large adsorbate island be formed at any coverage less than and including the saturation coverage for that structure. There may be several reasons for the formation of finite-size islands. Finite-size islands may occur because of kinetic limitations; that is, the large islands have not had sufficient time to grow at the expense of smaller ones. Substrate extended defects such as step edges may provide impenetrable barriers for the diffusion of adatoms.[2c,3] Each terrace separated by such steps then acts like an isolated thermodynamic subsystem. Finally, substrate defects may provide a contribution to the free energy of islands that makes small islands nucleated at such defects stable.

Low-energy electron diffraction (LEED) has been the major technique for investigations of island formation and growth,[4] including the study of islands on reconstructed clean[5,6] or adsorbate-covered surfaces.[1,2,7,8] However, reflection high-energy electron diffraction (RHEED) and grazing-angle x-ray diffraction can in principle also be used for these measurements. Information on the shape and size of the islands at various temperatures and coverages is contained in the angular distribution of the diffracted-beam intensity. In order to

extract the size distribution, to investigate critical nucleus sizes and to study the kinetics of growth of monolayer phases or the initial layer in epitaxy, it is important to be able to calculate the diffracted-beam intensity profile that is obtained for a distribution of overlayer islands.

A major aspect of the description of the diffracted-beam intensity profile from overlayer islands commensurately adsorbed on a crystalline substrate that has so far not been addressed analytically is the effect of interference between islands that may be spatially separated by quite a distance but that have some positional correlation. Commensurate adsorption implies that there is at minimum a correlation in the sites, i.e., all atoms sit in sites that occur at integral multiples of the lattice unit vectors. Therefore, no matter how far apart and how randomly the islands are formed on the surface, the positions of the islands, measured, for example, between centers, are still correlated through the periodicity of the substrate. Therefore the scattering from the islands is completely coherent, and one has to consider at all times the interference between the islands even if they are far apart. However, commensurate adsorption with a unit mesh greater than that of the substrate implies the existence of translational and perhaps also rotational antiphase boundaries.(9,10) Tracy and Blakely have suggested,(9) without rigorous proof, that if islands with a superlattice nucleate at random sites, the effect of interference between the islands (although they scatter coherently) is not important in the vicinity of the superlattice diffracted beams because of the antiphase relationship that must necessarily exist among such islands.

The superlattice beam intensity is then just the sum of the intensities scattered from the individual islands. This very important result has been used to determine the shape and size of the islands directly from the angular distribution of the superlattice beam intensity.(2c,2e,6,9-14) However, this result has not been proved rigorously and it is not clear how "random" the positions of the islands (recognizing that all atoms must sit on prescribed lattice sites provided by the substrate) must be in order for the result to hold. We shall address this problem here. We shall solve more generally the problem of the scattering of low-energy electrons from overlayer islands with a superlattice that have various degrees of positional correlation, using a rigorous mathematical treatment developed by Houston and Park.(15)

For a random distribution of islands, the separation of centers of two adjacent islands is an arbitrary integral multiple of the lattice constant $\underline{a}$. If the separations of any two neighboring islands are not arbitrary but have a limited range of values given by a distribution function $h(\underline{r})$, the islands are said to possess positional correlation. The function $h(\underline{r})$ is a set of δ-functions situated at integral multiples of $\underline{a}$, and it has appreciable value only in the vicinity of the average distance r_o. (Examples of the form of $h(\underline{r})$ will be given later). If the distribution of separations of second-neighbor islands, $h_2(\underline{r})$, is the self-convolution of $h(\underline{r})$, i.e., $h_2(\underline{r}) = h(\underline{r}) * h(\underline{r})$, then $h_2(\underline{r})$ is broader than $h(\underline{r})$. Similarly $h_3(\underline{r}) = h_2(\underline{r}) * h(\underline{r})$ is even broader than $h_2(\underline{r})$, and so on. It becomes more and more difficult to predict the positions of the islands at larger distances, and the islands are said to possess short-range positional correlation. On the other

hand, if the second-neighbor island-separation distribution function is given by $h_2(r) = h(r-r_o)$, the third-neighbor island separation distribution function by $h_3(\underline{r}) = h(\underline{r}-2r_o)$, and so on, then the width of the distribution function is identical for all $h_n(r)$. We shall refer to an overlayer with this island-separation distribution function as possessing long-range positional correlation.

We show rigorously that the result suggested by Tracy and Blakely[9] holds not only for a random distribution of islands, but also for islands having short-range positional correlation. We also calculate the interference for islands having long-range positional correlation. We point out that the contribution to the angular profile for this interference is distinguishable only under special circumstances of detector geometry.

In the next section we derive an expression for the diffracted-beam angular profile for islands adsorbed on a flat surface possessing short-range positional correlation, and in Sec. III for islands with long-range positional correlation. In Sec. IV the derivation is repeated for islands on a regularly stepped surface. Sec. V gives a brief summary.

II. Islands with Short-Range Positional Correlation

For simplicity we shall treat a one-dimensional problem. Consider small islands with lattice constant $2\underline{a}$ randomly arranged on a one-dimensional lattice with dimension $L\underline{a}$, where L is a large integer and a is the lattice constant. The islands are separated by regions of unoccupied sites or "sea". The substrate itself does not scatter, i.e., its scattering factor is assumed to be zero. The diffracted intensity can be written as[15]

$$I(x) = \overline{F^2}(x) + \bar{F}^2(x)\, F\,\{p(\underline{r})\} \quad , \qquad (1)$$

where $x = \underline{S} \cdot a$, the product of the diffraction vector $\underline{S}$ and the lattice constant $\underline{a}$, and $F\{\}$ stands for Fourier transform. The first term is simply the sum of the intensities scattered from the islands. The second term represents the interference between islands. For scattering from a set of commensurately adsorbed islands, the terms can be more explicity defined as follows: $F^2(x)$ is the structure factor of an individual island. For a collection of different-size islands with a doubly spaced arrangement of point scatterers the average of the square of the structure factor, $\overline{F^2}(x)$, is given by

$$\overline{F^2}(x) = \sum_{N=1}^{\infty} P(N) \sum_{\ell=0}^{2N} \exp(i\ell x) \sum_{\ell'=0}^{2N} \exp(-i\ell' x), \qquad (2)$$

where $\ell = 0, 2, 4 \ldots$ is an index that labels the ℓ'th site and $P(N)$ is the probability of finding an island of size $N2a$ containing $N+1$ scatterers whose scattering factor is assumed to be equal to one. The term $\bar{F}^2(x)$ is the square of the average of the amplitudes scattered from the island:

$$\bar{F}^2(x) = \Big[\sum_{N=1}^{\infty} P(N) \sum_{\ell=0}^{2N} \exp(i\ell x)\Big] \sum_{N'=1}^{\infty} P(N') \sum_{\ell'=0}^{2N'} \exp(-i\ell' x)\Big]. \quad (3)$$

The $F\,\{p(\underline{r})\}$ term in Eq. 1 is the Fourier transform of the autocorrelation function of island separations, i.e., $p(\underline{r})$ is the probability, given an island whose center is at the origin, of finding another island center a distance r away. The probability will be the same at $-\underline{r}$;

thus, $p(\underline{r}) = p(-\underline{r})$. $p(\underline{r})$ can be defined in terms of the nearest-neighbor island-center-position probability distribution function $h(\underline{r})$ mentioned earlier through the expression

$$p(\underline{r}) = \sum_{n=1}^{\infty} h_n(\underline{r}) + \sum_{n=1}^{\infty} h_n(-\underline{r}), \qquad (4)$$

where h_n is the n'th convolution product of $h(\underline{r})$,[15-17] i.e.,

$$h_n(\underline{r}) = h(\underline{r}) * \overset{n}{\ldots} * h(\underline{r}). \qquad (5)$$

Using the convolution theorem, Eq. (1) can be written in terms of Eqs. (4) and (5) as

$$I(x) = \overline{F}^2(x) + F^2(x) \left[\sum_{n=1}^{\infty} F^n \{h(\underline{r})\} + \sum_{n=1}^{\infty} F^n \{h(-\underline{r})\} \right], \qquad (6)$$

which can be summed to give

$$I(x) = \overline{F}^2(x) + F^2(x) \left[\frac{F\{h(\underline{r})\}}{1 - F\{h(\underline{r})\}} + \frac{F\{h(-\underline{r})\}}{1 - F\{h(-\underline{r})\}} \right] \qquad (7)$$

for $F\{h(\pm\underline{r})\} \neq 1$. A further simplication results if one defines

$$\rho e^{i\psi} \equiv F\{h(\underline{r})\} \quad . \qquad (8)$$

i.e.,

$$I(x) = \overline{F}^2(x) + F^2(x) \left[\frac{2\rho(\cos\psi - \rho)}{1 + \rho^2 - 2\rho\cos\psi} \right], \qquad (9)$$

where the quantities ρ and ψ will depend on the detailed form of $h(\underline{r})$.

To illustrate the case of islands with short-range positional correlation, we choose an initial form of the island position distribution function equal to

$$h(r) = \frac{1}{Q} \sum_{n=1}^{Q} \delta\,[r - (2M+n)\underline{a}], \qquad (10)$$

where Q is an integer ≥ 1 that represents the width of the distribution function $h(\underline{r})$, i.e., there is an equal probability (1/Q) of finding nearest-neighbor islands that are $(2M+1)\,\underline{a}$ ..., $(2M+Q)\underline{a}$ apart. This distribution function is shown in Fig. 1. The minimum island separation is given by $2M\underline{a}$, which is assumed to be larger than or equal to the largest island size. With this distribution function Q=1 always causes an antiphase relationship between nearest-neighbor islands. $h(\underline{r})$ is normalized such that the probability of finding nearest-neighbor islands separated by distances between $(2M+1)\underline{a}$ and $(2M+Q)\underline{a}$ is equal to one.

Taking the Fourier transform of Eq. (10) and solving for the quantities defined by Eq. (8) yields the following relationships:

$$\rho = \frac{1}{Q} \frac{\sin Q\, x/2}{\sin x/2} \qquad (11)$$

and

$$\psi = (2M + Q/2 + 1/2)\, x. \qquad (12)$$

These expressions can be evaluated for different beams. At the positions of the fundamental reflections [$x = h\pi$, (h = 0, 2, 4 ...)], $F(h(\underline{r})) = e^{i\phi} = 1$ for all values of M and Q, and $F(p(\underline{r}))$ is singular. Thus all atoms scatter in phase and there will therefore be strong interference effects at the positions of the fundamental reflections for all values of Q. At the positions of the superlattice reflections [for the doubly spaced structure we are assuming, $x = h\pi$ (h = 1, 3, 5 ...)], the evaluation of Eqs. (11) and (12) depends on the value of Q. For very large Q, i.e., for no correlations between island positions, $\rho \to 0$ and therefore $F(p(r)) \to 0$. There is thus no interference between islands and the total intensity is just the average intensity scattered from individual islands, given by the first term in Eq. (7). This is the result suggested by Tracy and Blakely[9] for islands with antiphase relationships nucleated "randomly" on a flat surface.

The effect of small or intermediate values of Q on the superlattice beam profile is less transparent. Small or intermediate values of Q imply that the probability of finding islands that are $(2M+1)a$... apart is large, i.e., there is positional correlation. We first note that at exactly a superlattice beam position, i.e., at $x = h\pi$, (h = 1,3,5 ...), $\cos\phi = (-1)^{1/2(Q+1)}$ when Q is odd and $\cos\phi = 0$ when Q is even, and $\rho = (-1)^{1/2(Q-1)}/Q$ when Q is odd and $\rho = 0$ when Q is even. In terms of our simple model, having Q even implies that two adjacent islands have an equal probability of having an inphase or an antiphase relationship. There is no net interference in this case, which is reflected in the fact that $\rho = 0$. Under these circumstances, according to Eq. (1), the intensity at the superlattice beam positions is

just the value of $\bar{F}^2(\pi)$. For small or intermediate even values of Q, the value of ρ remains small over many oscillations of $\cos\phi$ in the neighborhood of $x = h\pi$, (h=1,3,5...) and thus the second term in Eq. (4) makes a negligible contribution to the superlattice beam intensity. Hence, the diffracted intensity appears to originate from individual islands. This is the most general case for an overlayer with a superlattice that is adsorbed on a flat substrate.

If Q is odd, however, there is a larger probability that two adjacent islands have an antiphase relationship than an inphase relationship. This situation results, for example, when islands grow to the size where they can coalesce. Because only islands that do not have an antiphase relationship can coalesce, there will be an excess of antiphase boundaries. This generally causes some *destructive interference* at the superlattice beam positions, i.e.,

$$F(p(\underline{r})) = -\left(\frac{2}{Q+1}\right) \qquad (13)$$

and

$$I(\pi) = \bar{F}^2(\pi) - \left(\frac{2}{Q+1}\right) F^2(\pi). \qquad (14)$$

Eqs. (13) and (14) show that if Q is odd, a broad distribution of island separations (Q large) washes out the interference and the superlattice beams look like they result from the average island structure factor, as for Q even. Interference effects will be significant only for small Q with Q odd. In this situation, the well-known spot splitting results.

As an illustration of the effect of Q on the superlattice beam profile we calculate the changes in the angular profile of the first-order superlattice reflection caused by the contribution of the interference term [second term in Eq. (9)] for Q = 2, 3, 10, and 11 for a constant island size of $2Na = 20a$ and center separations $2Ma = 40a$. A comparison of profiles with and without the interference term for those cases is shown in Figs. 2a-d. For small even Q, the interference changes the profile only slightly, with the width remaining approximately unchanged. This agrees qualitatively with the result of recently published Monte-Carlo simulations.[14] On the other hand, for small, odd Q the beam profile takes on the classical appearance of the split beam common to antiphase behavior. For intermediate Q, the effect is less pronounced.

The above results also hold for systems with overlayer unit mesh equal to $q\underline{a}$, where q is an integer > 2, (e.g. 3x1, 4x2). One merely redefines

$$h(\underline{r}) = \frac{1}{Q}\sum_{n=1}^{Q} \delta\,[\underline{r} - (qM + n)\underline{a}]. \qquad (15)$$

For this island separation distribution function

$$\nu = \frac{1}{Q}\frac{\sin Q\,x/2}{\sin x/2} \qquad (16)$$

and

$$\psi = (qM + \frac{Q}{2} + \frac{1}{2})x. \qquad (17)$$

In the model discussed in this section there is no long-range correlation between the positions of the islands (except that atoms must sit at lattice sites and thus island separations must be integral multiples of $\underline{a}$). The value of $p(\underline{r})$ at the lattice positions approaches a constant for large $\underline{r}$. We shall now consider a model that has long-range correlation in island positions, but maintains the possibility of an antiphase relationship between the islands.

III. Islands with Long-Range Positional Correlation

In the simple model illustrated by Eq. (10), the ability to locate an island boundary decreases with the number of intervening islands, which is physically realistic for islands adsorbed on a flat substrate. However, it is possible to have an experimental condition in which island positions are highly correlated over long distances. *An example* of this would be commensurate adsorption on (or reconstruction of) a regularly stepped surface.[5,6,10,18] To illustrate the effect of this type of adsorption on diffracted beam profiles we shall first consider islands with double the substrate periodicity, i.e., q=2, adsorbed on a flat substrate with long-range positional correlation. We model the problem by making a direct assumption of the form of the autocorrelation function that includes this positional correlation, i.e.,

$$p(\underline{r}) = \frac{1}{4}\sum_{\substack{n=-R+1\\ n\neq 0}}^{R-1} \{2\delta(\underline{r}-nM2\underline{a}) + \delta[\underline{r}-(nM2\underline{a}+\underline{a})) + \delta[\underline{r}-(nM2\underline{a}-\underline{a})]\}. \qquad (18)$$

where $M2a$ is the average periodicity of the island positions and $2R$ is the total number of islands. $p(r)$ is shown in Fig. 3. The sizes of the islands are obviously smaller than $M2a$. The intensity resulting from this positional correlation can be expressed as

$$F(p(r)) = \frac{1}{2}(1 + \cos x) \sum_{\substack{n=-R+1 \\ n\neq 0}}^{R-1} \exp[i2nMx]$$

$$= (A-1)\cos^2 x/2, \qquad (19)$$

where

$$A = \frac{\sin\ (2R-1)Mx}{\sin\ Mx}. \qquad (20)$$

A has peaks at $x = \frac{h'\pi}{M}$, ($h'=0,1,2\ldots$), reflecting the average periodicity of the island positions. The height and width of the peaks are proportional to $2R-1$ and $\frac{1}{(2R-1)M}$ respectively. As the number of islands $R \to \infty$, A can be represented by a set of delta functions with an area proportional to $\frac{1}{M}$, i.e.,

$$A = \frac{\pi}{M} \sum_{h'} \delta\left(x - \frac{h'\pi}{M}\right). \qquad (21)$$

The area under the δ-function is a constant and does not increase as the number of islands $R \to \infty$. The term $\cos^2 \frac{x}{2}$ is the result of the interference originating *in the antiphase relationship among the* islands. The term is small around a superlattice beam, indicating cancellation of phases due to the antiphase relationship.

$F(p(r))$ is everywhere zero except at exactly $x = \frac{h'\pi}{M}$, where the function goes to infinity. At $x = \frac{h'\pi}{M}$, the integrated intensity of $F(p(r))$ may still be small and negligible because $\cos^2 \frac{x}{2}$ is close to zero. *(Recall that the area under the δ-function remains* constant as $R \to \infty$). This is seen by integrating the intensity profile over a detector of width ϵ. Assuming that ϵ is small, the intensity integrated over ϵ at a particular value of $h' = Mh+1$ near the superlattice beam position, $x = h\pi + \frac{\pi}{M}$, ($h=1,3,5\ldots$), can be written approximately as

$$I_{\text{integrated}}\left(x = h\pi + \frac{\pi}{M}\right) = \bar{F}^2 \epsilon + F^2 \left[\sin^2 \frac{\pi}{2M}\right] \int_{h\pi+\frac{\pi}{M}-\frac{\epsilon}{2}}^{h\pi+\frac{\pi}{M}+\frac{\epsilon}{2}} \left[\frac{\pi}{M}\ \delta\left[x-\left(h\pi+\frac{\pi}{M}\right)\right]-1\right] dx$$

$$= \bar{F}^2 \epsilon + F^2 \left[\sin^2 \frac{\pi}{2M}\right]\left[\frac{\pi}{M} - \epsilon\right]. \qquad (22)$$

As $\epsilon \to 0$, the second term dominates and the intensity profile $I(x)$ has a spike at $x = h\pi + \frac{\pi}{M}$. Figure 4 is a plot of the superlattice beam profile for $M = 20$. The island size has been assumed to be identical for all islands and equal to $M2a = Ma$, i.e., half the size of the average periodicity of the island positions. The spikes, *which are a reflection of the periodicity of the island positions,* sit on the average intensity scattered from the individual islands, $\bar{F}^2(x)$. The spacing of the spikes is $1/M$ of the spacing of the main diffraction peaks. There is no spike at the center of the superlattice beams, $x = h\pi (h=1,3,5\ldots)$, because $\cos^2 \frac{x}{2} = 0$ at these points.

As ϵ gets larger, the relative contribution of the second term gets smaller. The interference effect approaches zero as $\epsilon \to \frac{\pi}{M}$. For islands of equal size $M2a$, the ratio of the second to the first term in Eq. (22) as a function of ϵ is given by

$$Z = [(\frac{\pi}{M} - \epsilon)/\epsilon] \sin^2 \frac{\pi}{2M}. \qquad (23)$$

This is plotted in Fig. 5 for a repeat distance $M = 20$. This figure shows that the contribution of the interference is negligible except for an extremely small detector width. Z is only 5% for a detector with a width of 1/10 of the period $\frac{\pi}{M}$ in reciprocal space. Typical separations of diffraction spots are of the order of 10° or less. For an island center spacing of $M = 20$, this translates into a detector angular aperture of 1/20°, much less than is commonly used. For larger M, even smaller detectors are required to observe an effect due to antiphase boundaries for islands with long-range positional correlation. Thus for practical purposes the measured superlattice beam profile is just that which would be obtained from independently scattering islands.

The above result is easily generalized to overlayer islands of unit mesh qa for $q > 2$. The autocorrelation function can be written as

$$p(\underline{r}) = \frac{1}{q^2} \sum_{n=-(R-1)}^{R-1} \sum_{m=1}^{q-1} [(q-m)\{\delta[\underline{r}-(nMq-m)\underline{a}] + \delta[\underline{r}-(nMq+m)\underline{a}]\} + q\delta[\underline{r}-nMq\underline{a}]] \qquad (24)$$

and

$$I(p(\underline{r})) = \frac{1}{q^2} \frac{\sin^2 qx/2}{\sin^2 x/2} \quad (A-1), \qquad (25)$$

where

$$A \quad \frac{2\pi}{qM} \sum_{h'} \delta\left(x - \frac{2h'\pi}{qM}\right). \qquad (26)$$

Clearly the problem can also be generalized to include a distribution of island sizes, rather than the single size assumed here.

IV. <u>Islands on a Regularly Stepped Surface</u>

Commensurate adsorption of an overlayer on a regularly stepped surface is an example of a situation in which islands may have long-range positional correlation. The scattering of low-energy electrons from overlayer islands on stepped surfaces has been considered for two special cases.[6,10,18] Here we shall consider a general treatment based on a model similar to that of Section III.

We assume that the overlayer island positions possess long-range correlation through a regularly stepped substrate. Eq. (18) can be modified to give

$$p(\underline{r}) = \frac{1}{4} \sum_{\substack{n=-R+1 \\ n \neq 0}}^{R-1} [2\,\delta\{\underline{r}-[(n2M+\tfrac{1}{2})\underline{a}+t\underline{\ell}]\} + \delta\{\underline{r}-[(n2M+\tfrac{3}{2})\underline{a}+t\underline{\ell}]\} + \delta\{\underline{r}-[(n2M-\tfrac{1}{2})\underline{a}+t\underline{\ell}]\}], \qquad (27)$$

where $2Ma$ is the terrace width, c is the unit vector normal to the surface and t is the step height. There is an $\frac{a}{2}$ lateral displacement at the step edge. On each terrace the adsorption may be into one site or into the translationally displaced site, which leads to an antiphase relationship among the islands. Figure 6 shows schematically a stepped surface without the overlayer. The autocorrelation function $p(\underline{r})$ defined in Eq. (27) leads to the same results given by Eq. (19), except that A is now defined by

$$A = \sum_{h'}^{\frac{M}{M+\frac{1}{4}}} \delta\left[x - \frac{h'\pi - S_{\perp}t}{M + 1/4}\right] . \qquad (28)$$

where $S_{\perp}$ is the momentum transfer perpendicular to the surface. Therefore, the spikes occur at

$$x = \frac{h'\pi - S_{\perp}t}{M + 1/4} , \qquad (29)$$

which depends on energy through $S_{\perp}$. As before, spikes will occur at particular values of x, but unless the detector aperture is very narrow, the contribution to the intensity from the interference terms is negligibly small and the measured superlattice beam profile is just that which would result from independently scattering islands adsorbed on a flat surface. This, in fact, has been observed experimentally.[5,6]

If the island positions are perfectly periodic, i.e., if the translational displacement between islands on different terraces is not allowed, the autocorrelation function is given by the first term in the summation in Eq. (27). This could happen if the islands are all required to nucleate at step edges rather than randomly on the terraces. In this case there is no random antiphase relationship

among the islands and the superlattice beams will show the familiar phenomenon associated with regularly stepped surfaces and observed in the fundamental reflections[19], namely the alternate appearance of single and double ("split") reflections as the energy of the incident electrons is changed.

V. Summary

We have calculated the angular distribution of LEED intensity from commensurately adsorbed overlayer islands with a unit mesh larger than that of the substrate that possess positional correlation. In the calculations, the positional correlation among the islands is represented by a set of δ-functions. The intensity is evaluated using the method developed by Houston and Park.[15,16] Closed-form solutions are obtained for models that produce short-range and long-range correlation for the island positions but maintain an antiphase relationship[9] among the islands. The results agree with previous qualitative conclusions. For islands with short-range positional correlation and with a random antiphase relationship, the shape of the superlattice beam profiles is affected only slightly (depending on the degree of correlation) by the interference caused by the antiphase relationship. The effect is in any case negligible on the scale of measurement accuracy. If the antiphase relationship is not random but regular, the familiar beam splitting results. For islands with long-range positional correlation and with a random antiphase relationship, the superlattice beam profile shows sharp spikes sitting on top of the intensity scattered from individual islands. These spikes are weak and give a negligible contribution when a finite-size

detector is used. For a regular antiphase relationship, beam splitting results again, of course.

Calculations are also presented for islands on a regularly stepped surface, which is a possible physical manifestation of islands with long-range positional correlation.

The derivations have in all cases assumed the kinematic approximation for scattering and substrate atomic scattering factors that are equal to zero. The results also hold if we assume that the substrate atomic scattering factor is not zero, at least within the kinematic approximation, because in this approximation the substrate contributes no amplitude at the superlattice beam positions. Frequently, however, overlayers form a (1x1) structure, i.e., no superlattice exists. In that case, because there are only integral-order beams, the effect of the substrate scattering is important in the derivation of the diffracted beam shape and the diffraction problem is more complicated than the results presented here. If the adsorbate and substrate atoms are identical, the system is identical to a surface with alternately up and down monatomic steps, a problem that has been discussed extensively in the past[19,20]. If the adsorbate and substrate atoms are different, then in the calculation of the beam shape the effect of the phase difference in the atomic scattering factors must also be considered. We are making calculations to simulate this situation.[21]

The results presented here are independent of the technique used to measure the diffracted beam profiles. In essence, we have described a distribution of intensity in reciprocal space and the

conclusions apply to any technique that is sensitive to a single atomic layer. These techniques include low-energy electron diffraction (LEED), reflection high-energy electron diffraction (RHEED), and grazing-angle x-ray diffraction.

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Figure Captions

Fig. 1. The distribution function $h(r)$, given by Eq. (8), of island positions for a model of islands with short-range positional correlation.

Fig. 2. Angular profiles calculated for the first-order superlattice reflection (h=1) from islands with a unit mesh twice that of the substrate and having short-range positional correlation, for several values of the separation Q of islands (i.e., the coverage or equivalently the island density is changing). The intensity is plotted as a function of $x = \underline{S}\cdot\underline{a}$ around the value $x = \pi$. a) $Q = 2$, b) $Q = 3$, c) $Q = 10$, d) $Q = 11$. Solid curves: without interference term due to antiphase boundaries. Dashed curves: with interference. Large Q corresponds to large island separation and hence low coverage. Even Q implies an equal probability of inphase and antiphase boundaries. The profiles depend only weakly on interference effects. Odd Q implies a predominance of antiphase boundaries. Characteristic spot splitting occurs. This splitting is more pronounced for small Q (small island separation) than for large Q. The islands are all the same size, $2N = 20$ lattice constants. The subsidiary maxima are caused by the choice of a single island size. They disappear if a distribution of sizes is chosen.

Fig. 3 Example of an autocorrelation function of islands with long-range positional correlation. $p(\underline{r})$ continues to infinity. $p(\underline{r})$ is given by Eq. (14).

Fig. 4 Schematic angular profile of an odd-order reflection from a distribution of islands with unit mesh twice that of the substrate and having long-range positional correlation. The islands are the same size $2N = 20$ lattice constants. The periodicity of the island positions is 40 lattice constants. The subsidiary maxima are caused by the choice of a single island size. The delta functions at $\pm\pi/M$ are due to interference between the islands. The strength of these δ-functions depends on the separation of islands and their size. δ-functions at $\pm\frac{3\pi}{M}$, ... are weak and not shown. Because of complete destructive interference, there is no δ function at the center of the profile.

Fig. 5 Ratio of the sum of integrated intensities from individual islands (first term in Eq. (22)) and the integrated intensity due to interference between correlated islands (second term in Eq. (22)) as a function of the detector size ϵ. ϵ is given in reciprocal lattice units. $\epsilon = 2\pi/M$ corresponds to a detector angular aperture that would just include the center of the diffracted beam and the first delta function spike for islands with long-range positional correlation that have a unit mesh twice that of the substrate.

Fig. 6 Schematic diagram of a stepped surface for a solid with AB or ABC stacking. The autocorrelation function for overlayer island positions on such a stepped surface is given by Eq. (27). a is the lattice constant and t is the step height.

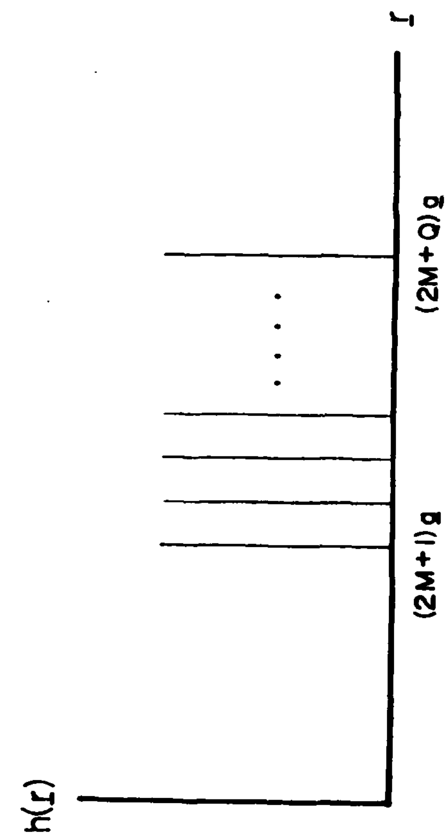
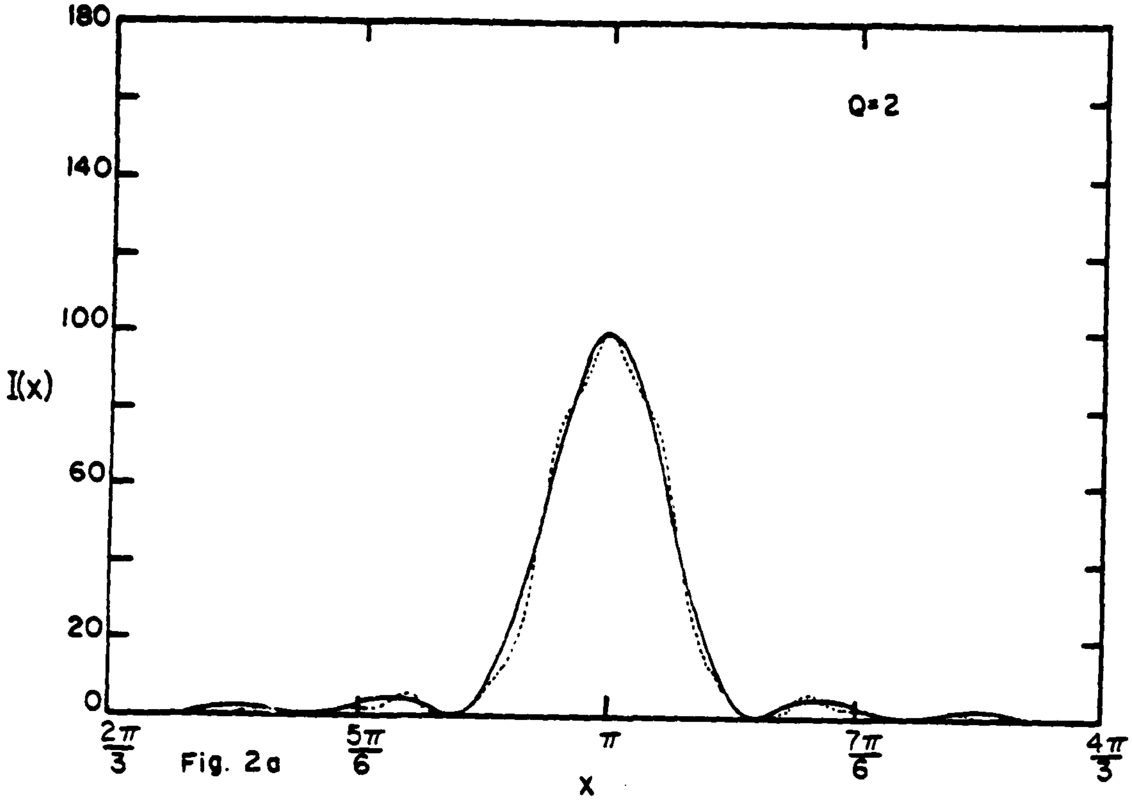
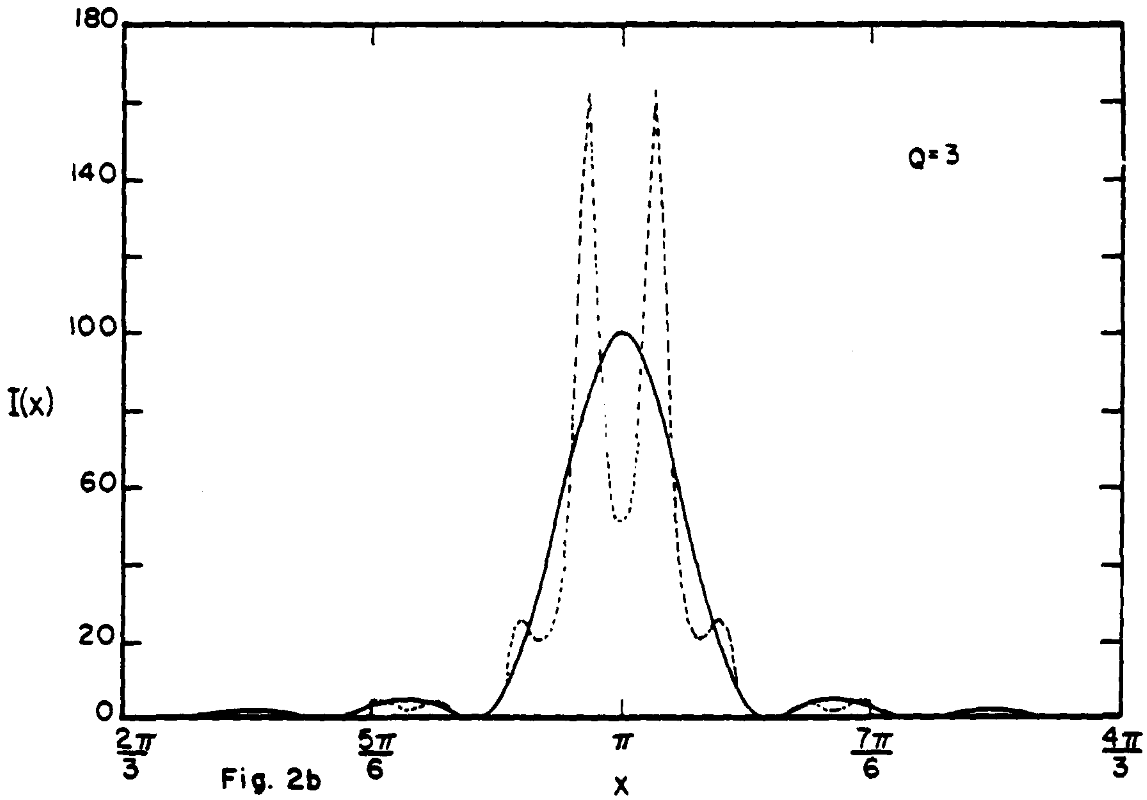


Fig. 1



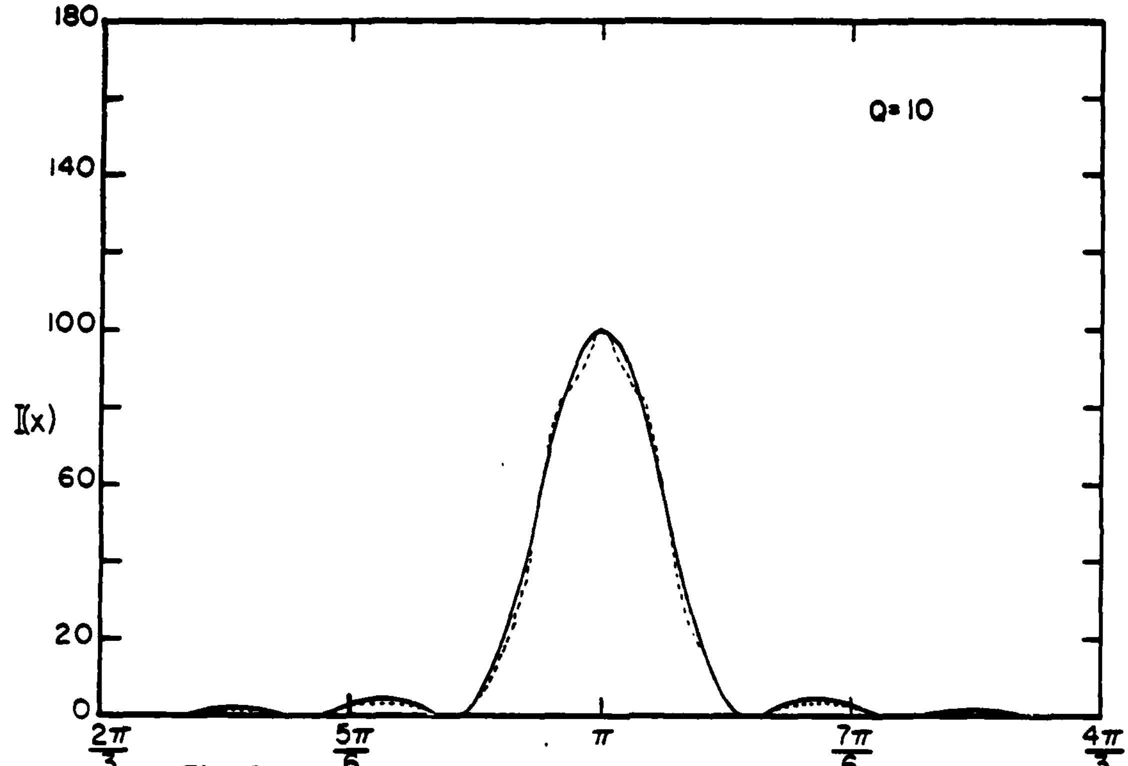
180
140
100
60
20
0
I(x)
Q=2
$\frac{2\pi}{3}$
$\frac{5\pi}{6}$
π
$\frac{7\pi}{6}$
$\frac{4\pi}{3}$
X

Fig. 2a



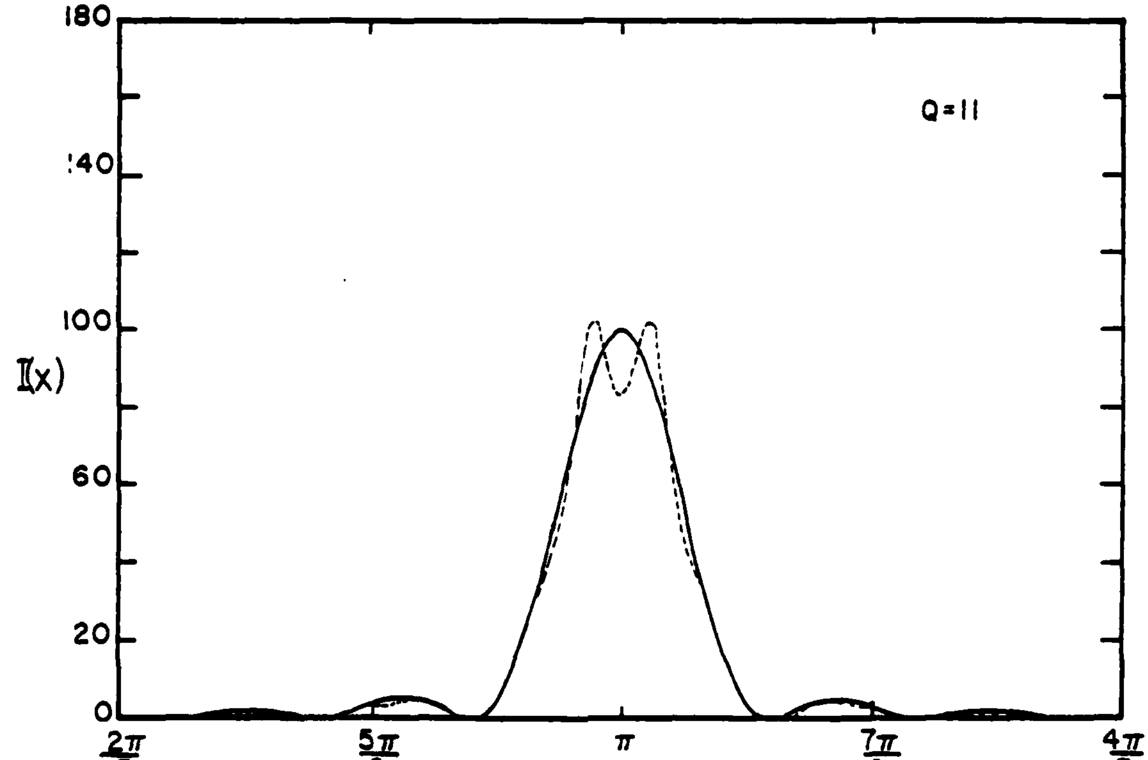
180
140
100
60
20
0
I(x)
Q=3
$\frac{2\pi}{3}$
$\frac{5\pi}{6}$
π
$\frac{7\pi}{6}$
$\frac{4\pi}{3}$
X

Fig. 2b



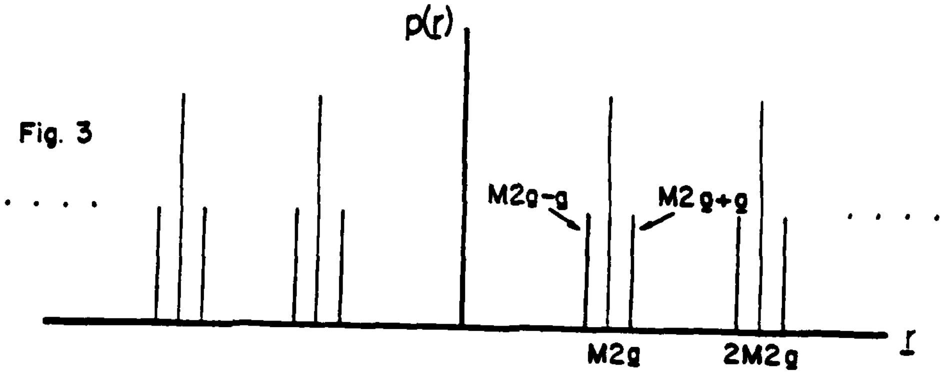
180
140
100
60
20
0
Q=10
I(x)
2π/3
5π/6
π
7π/6
4π/3
X

Fig. 2c



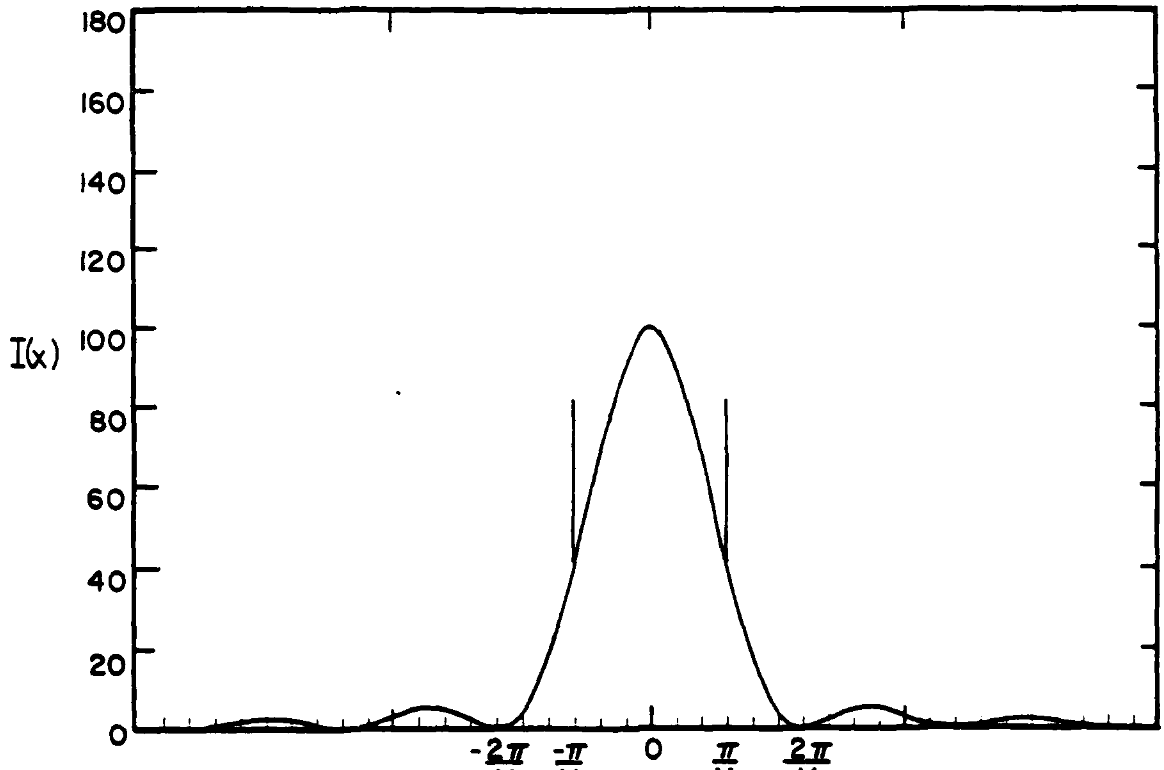
180
140
100
60
20
0
Q=11
I(x)
2π/3
5π/6
π
7π/6
4π/3
X

Fig. 2d.



p(r)
M2g-g
M2g+g
M2g
2M2g
r

Fig. 3



180
160
140
120
100
80
60
40
20
0
I(x)
-2π/M
-π/M
0
π/M
2π/M
x-hπ

Fig. 4

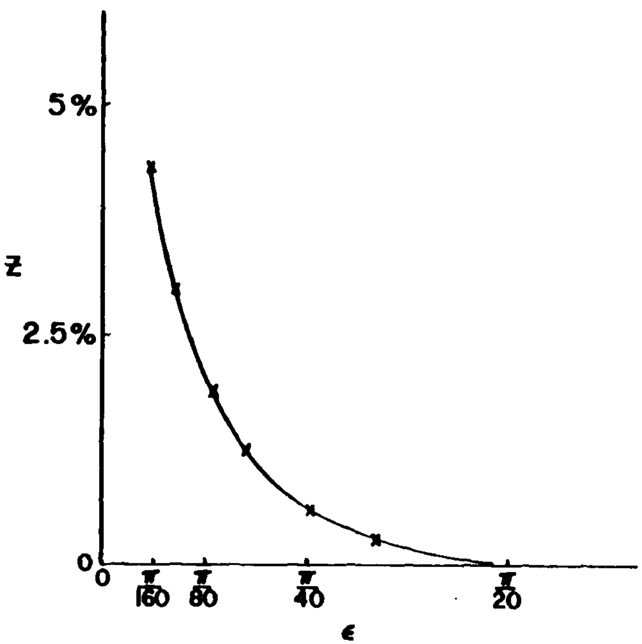
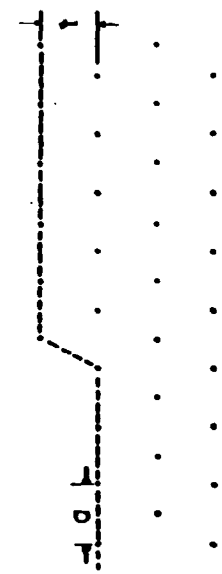


Fig. 5



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